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16. ABSTRACT

Under its Indoor Air Quality research program, the U. S. Environmental Protection Agency (EPA) has developed several new air monitoring systems for assessment of nonoccupational exposures to chemicals found primarily in indoor air. Integrating and real-time (continuous) instruments for nitrogen dioxide, carbon monoxide, formaldehyde and a broad spectrum of volatile organic chemicals have been designed and field tested for indoor air and/or personal exposure monitoring. Samplers capable of collecting sufficient quantities of semivolatile organic chemicals for ultra-trace analysis and bioassay, which are also quiet and unobtrusive, have been developed and evaluated.

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DEVELOPMENT AND EVALUATION OF INSTRUMENTATION FOR MEASUREMENT OF INDOOR AIR QUALITY

R. G. LEWIS

Methods Research Branch, Methods Research and Development Division, Atmospheric Research and Exposure Assessment Laboratory, U. S. Environmental Protection Agency, Research Triangle Park, North Carolina 27711 (U.S.A.)

SUMMARY

Under its Indoor Air Quality research program, the U. S. Environmental Protection Agency (EPA) has developed several new air monitoring systems for assessment of nonoccupational exposures to chemicals found primarily in indoor air. Integrating and real-time (continuous) instruments for nitrogen dioxide, carbon monoxide, formaldehyde and a broad spectrum of volatile organic chemicals have been designed and field tested for indoor air and/or personal exposure monitoring. Samplers capable of collecting sufficient quantities of semivolatile organic chemicals for ultra-trace analysis and bioassay, which are also quiet and unobtrusive, have been developed and evaluated.

1. INTRODUCTION

The typical resident of the United States and western European countries spends 95% or more of the time within enclosed air spaces in the home, workplace, public buildings, or transit vehicles (1). Most air pollutants have been shown to be present in indoor environments at concentrations that are usually several times to one-hundred times higher than in the surrounding outdoor air (2). Consequently, indoor air quality should be of considerably greater concern in most cases than outdoor air quality with respect to chronic respiratory exposure to potentially harmful toxic substances.

The U. S. government recognized the need to assess indoor air pollutants in 1982 when funding appropriations to the Environmental Protection agency (EPA) were authorized by the Congress. In 1986, the Radon Gas and Indoor Air Quality Act mandated that EPA establish a research program on all aspects of indoor air quality. One of the earliest needs recognized was for adequate indoor air monitoring methodologies (3).

Air monitors suitable for use in nonoccupational indoor environments must be unobtrusive, easily portable, highly sensitive, rugged and affordable. Most samplers used for outdoor air measurements are too large, noisy, and have flow rates too high for indoor application. Monitors designed for industrial hygiene purposes generally lack the sensitivity required for general population exposure assessment. Pollutants of particular concern for indoor air measurement are volatile organic chemicals (VOCs); pesticides; polynuclear aromatic hydrocarbons (PAHs); radon and its progeny; oxides of nitrogen and carbon; and respirable and inhalable particles. With the exception of radon, most of these pollutants arise from internal sources such as combustion from heating and cooking, emissions from building materials and furnishings, fumes from attached garages, or human activities such as smoking, use of pesticides and disinfectant products and bathing. Significant recent progress has been made in the development of sensitive monitors for several of these pollutants.

2. VOLATILE ORGANIC CHEMICALS

Most large monitoring studies of non-polar volatile organic chemicals (VOCs) in indoor air have involved collection on activated charcoal (4,5,6) or Tenax®-GC (7,8). While Tenax is the preferable sorbent since it can be thermally desorbed to permit much lower limits of detection, both methods are subject to problems with artifact formation that may lead to false-positive identifications. One technique that is essentially free of artifact problems, was recently developed by EPA (9,10). The method employs whole-air collection in evaculated stainless-steel canisters, the interiors of which have been passivated by an electropolishing procedure known as SUMMA®-polishing. Air may be sampled into these canisters at controlled flow rates either by use of a critical orifice or mass-flow controller without the aid of a pump, or a pump may be used to fill the canisters to pressures up to several atmospheres. An aliquot (usually 200 mL) of the canister contents is cryofocused into a gas chromatograph for analysis with a mass selective detector (MSD) or traditional GC detector. Detection limits are generally below 1 $\mu g/m^3$ when an MSD is used, and may be much lower for certain VOCs using other detectors. A distinct advantage over thermally-desorbed sorbent sampling is the ability to perform repeated analyses on the same or different analytical systems. Non-polar VOCs with vapor pressures as low as 10^{-2} kPa at 20° C and polar VOCs as reactive as ethylene oxide have been shown to be stable inside the canisters for periods of one to several weeks. However, when MSD analysis is performed, the sample aliquot is normally passed through a perfluorinated ionomer membrane (Nafion® tubing) to remove co-collected water. This process also effectively removes many of the polar VOCs. Chemical ionization MS using water vapor or other chemicals as the reagent gas is currently under investigation to permit *in situ* analysis of polar VOCs with the ion trap detector.

In one sampler design used successfully by EPA, 6-L canisters are filled passively by means of mass-flow controllers. At 3-4 cm^3/min, *ca.* 5.5-L of air can be collected over a 24-h period. A timer and solenoid-controlled valving system has also been designed to permit intermittent sampling with this system for periods up to one week (10). With pump-driven samplers, *ca.* 14 L of air can be sampled into the 6-L canister at 11 cm^3/min over 24 h. Several U.S. manufacturers have recently introduced this type of sampler based on EPA designs. Some of these are compact (*e.g.*, 30 x 56 x 48-cm, 23 kg) and are essentially quiet in operation, making it well suited for indoor sampling. Since they are also weatherized, the same sampler may be used outside.

A variation of the canister sampler is a 12-syringe sampler designed by EPA in cooperation with its manufacturer. It is portable, battery-operated, and can be programmed to consecutively fill the SUMMA® polished syringes (150 cm^3 each) at rates ranging from 2 to 90 min per syringe (11). Analysis by GC/MSD is fully automated to analyze sequentially the contents of each syringe with a sensitivity less than 1 $\mu g/m^3$ for most VOCs. The sampler has been used to determine temporal profiles of VOCs in buildings and motor vehicles.

3. SEMIVOLATILE ORGANIC COMPOUNDS

3.1 Pesticides

The methodology used most widely in the U.S. for monitoring pesticides in indoor air was developed by EPA in 1982 (12). It has recently been used in a large nonoccupational exposure study involving about 270 households to determine airborne concentrations of over 30 pesticides (12,13,14) and has been adopted by ASTM in the U.S. (16) and WHO in Europe (7). Small personal pumps are used to sample air at 3.8 L/min through a cylinder of polyurethane foam 22-mm o.d. x 7.6-cm long. The 5.5 m^3 volume of air sampled in 24 h provides limits of detection ranging from 1 to 100 ng/m^3 depending on the compound and analytical finish (GC/MS or GC/ECD).

3.2 Polynuclear Aroma

Polyurethane foam
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for nitro- and oxy-PAHs
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4. INORGANIC GASES

4.1 Nitrogen Dioxide

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4.2 Carbon Monoxide

The most common me
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electrochemical monitor

3.2 Polynuclear Aromatic Compounds

Polyurethane foam (PUF) has also been used extensively to sample for polynuclear aromatic hydrocarbons (PAHs) in air. Most outdoor sampling has utilized either a modified Hi-Vol (18) or PS-1 sampler (19), both of which pull air through a particle filter followed by a PUF trap to collect PAH vapors. The latter sampler, which can achieve flow rates up to 250 L/min, was modified by EPA for indoor use by housing the pumping system in an acoustically-insulated cabinet (20). This sampler is very quiet in operation and has been successfully used in several multi-home studies. It provides a sufficient sample volume in 12 to 24 h for in-depth GC/MS analysis as well as mutagenicity testing. However, it is large, and the flow rate is too high for use in smaller enclosed areas. Consequently, a lower flow sampler (15 to 20 L/min) has been designed with similar features (21). Air is drawn through a 47-mm circular quartz-fiber filter and subsequently through a glass cartridge containing 60 g of Amberlite® XAD-2 resin. When evaluated for continuous 24-h periods (150 m^3 sample volume) and in an intermittent mode for 7 days (190 m^3 sample volume), the collection efficiency of the sampler is 90% or better for 29 PAHs with two to six rings. Detection limits using electron ionization GC/MS for nonpolar and negative chemical ionization GC/MS for nitro- and oxy-PAHs were 0.1 ng/m^3 or lower. When XAD-4 resin is used, nicotine is also efficiently collected (21).

4. INORGANIC GASES

4.1 Nitrogen Dioxide

Most indoor air measurements of NO_2 have been performed with Palmes diffusion tubes (22). With a sampling rate of 1 cm^3/min and a normal sensitivity limit of 0.5 mg-h/m^3, exposure periods of 5 to 7 days are usually required. EPA recognized the need for a more sensitive passive sampler for NO_2 that would permit short-term integrated measurements to assess household exposures. To meet this need, a high-efficiency passive sampling device (PSD) developed for VOCs (23) was adapted to collect NO_2 (24). The small PSD consists of a series of diffusion barriers placed on either side of glass fiber filters treated with triethanolamine. The effective sampling rate is 154 cm^3/min and sensitivity 25 µg-hr/m^3 when ion chromatography is used for analysis. Indoor air studies demonstrated that the PSD could easily determine NO_2 levels ranging from 24 to 140 $µg/m^3$ with 12- to 24-h exposures. During cooking with stoves fueled with natural gas, exposure periods of 20 to 60 min were adequate (observed concentrations 150-520 $µg/m^3$).

Since brief exposures to high concentrations of NO_2 are believed to present a cumulative health hazard, especially to children, a small continuous monitor was also needed. Initial developmental efforts sponsored by EPA resulted in a commercial chemiluminescence monitor, the Scintrex Luminox® LMA3, with a sensitivity of 1 ppbv and a response time of 10 sec (24). The size (38 x 20 x 22 cm) and weight (7 kg) of this monitor and its battery-operation capability make it ideally suited for indoor use. EPA is currently developing a small or real-time NO_2 monitor based on electrochemical detection that can be also used for personal monitoring. A prototype device measuring 6.3 x 13 x 20 cm and weighing 1.2 kg has a detection limit of 5 ppbv and a response time of 30 sec (26).

4.2 Carbon Monoxide

The most common method for CO in air is based on continuous measurement by non-dispersive infrared (NDIR) spectroscopic detection. While sensitive enough, NDIR instruments are much too bulky and complicated for indoor air quality monitoring. To address indoor and personal monitoring needs, a small electrochemical monitor called the "COED" was developed by EPA under contract

(27). The monitor, which measures 15 x 19 x 13 cm and weighs less than 1 kg contains a sensing cell with a solid polymer electrolyte and an internal microprocessor data logger that records and stores up to 100 readings. The monitor has been successfully used in two large field studies (28,29). The EPA-developed gas filter correlation monitor, which also determines water vapor, is also suitable for indoor area monitoring (30).

4.3 Radon

Satisfactory instrumentation for determining indoor air levels of radon and radon progeny is available. The major problem currently is the proper use of a measurement device to provide accurate values. The EPA has recently published interim guidelines for the use of integrating, continuous, and grab sampling methods (31). Charcoal canisters (8 or 10 cm in diameter) that are analyzed by gamma ray scintillation are useful for 1- to 7-day sampling. Alpha track detectors may be used to obtain integrated measurements over periods of 1 to 12 months. Recently developed electret monitors can be produced for short-term to long-term integrative sampling (32). The monitor employs an electrostatically-charged disk that experiences a reduction in surface charge proportional to the amount of radon exposure. EPA has also instituted a National Radon Measurement Proficiency program, under which radon monitors submitted by manufacturers are evaluated and listed.

5. FORMALDEHYDE

Many methods have been applied for determining formaldehyde in indoor air. The most popular have been impingement into sodium bisulfite solutions followed by colormetric analysis using chromotropic acid (NIOSH method 3500) or deployment of commercially-made PSDs, such as the 3M 3750 monitor. Most of these methods are sensitive enough only for workplace atmospheres, where concentrations are relatively high. An EPA-developed active method employs a silica gel cartridge coated with 2,4-dinitrophenylhydrazine (2,4-DNPH) and has a detection limit of less than 2 ppbv-h (33). Air is pulled through a small SepPAK® cartridge treated with the reagent at 0.5 to 1.5 L/min for 1 to 24 h. Analysis by HPLC can determine concentrations of formaldehyde and 14 other aldehydes, as well as acetone. The PSD described in Section 4.1, has also been adapted to determine aldehydes by the use of 2,4-DNPH-coated filters (24). The PSD samples at 103 cm^3/min and has about one-tenth the sensitivity of the active method. EPA has recently modified the CEA TGM-555 continuous monitor for formaldehyde to improve its stability and sensitivity (34). The monitor is highly portable (50 x 40 x 1.8 cm, 13.6 kg) and can operate on rechargeable batteries for up to 12 h. By changing the pararosaniline chemistry (eliminating the sodium tetrachloromercurate) and reconfiguring the liquid flow release, the monitor can continuously monitor formaldehyde at low levels (10 to 100 ppbv) with high accuracy and precision.

6. RESPIRABLE AND INHALABLE PARTICLES

As in the previous cases, particle samplers for outdoor ambient air monitoring are large, noisy, and cumbersome. Some continuous inhaled particle (IP) monitors are available (e.g., the piezobalance), but they also are too large, too inaccurate, or too complex for indoor air quality monitoring. The smaller commercial monitors have poor sensitivity and/or unknown validity at the low particulate concentrations found in nonoccupational environments. To address the need for an indoor air monitor, EPA supported the development by the National Institute for Standards and Technology (NIST) of a small, dual channel sampler (35) that collects 0 to 2.5 μm (respirable) and a 2.5 to 10 μm (inhalable) size fractions, operates at 6 L/min, and is powered by a battery pack. The particle separation between the channels is geometric and achieved by using an 8-mm Nuclepore® filter followed by an absolute filter. The sampler measures 10 x 10 x 18 cm and weighs 1.6 kg. It can operate

continuously for 20 to 6
recently been developed
holder for a 37 mm Teflo
(2.5 and 10 μm) that can
vide an air flow of 4 L/
can be collected in 24 h

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continuously for 20 to 60 h. A small personal particulate sampler has also recently been developed by EPA (36). It consists of a cylindrical metal holder for a 37 mm Teflon filter (2 x 4 cm) with two interchangeable orifices (2.5 and 10 μm) that can be worn on the person and a personal pump to provide an air flow of 4 L/min through the device. Sufficient particulate matter can be collected in 24 h for gravimetric and X-ray fluorescence analysis.

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SUMMARY

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INTRODUCTION

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